International Journal of Multiphase Flow 60 (2014) 30–39

Contents lists available at ScienceDirect

International Journal of Multiphase Flow

journal homepage: www.elsevier.com/locate/ijmulflow

Air–water mass transfer mechanism due to the impingement of a single liquid drop on the air–water interface

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ARTICLE INFO

Article history:
Received 18 August 2013
Received in revised form 15 November 2013
Accepted 23 November 2013
Available online 5 December 2013

Keywords:
Surface exchange process
Mass transfer coefficient
Gas-liquid interface
Surface renewal theory
Vortex ring
Rainfall

ABSTRACT

The mass transfer mechanism across the air–water interface due to the impingement of a single liquid drop was investigated through laboratory experiments using particle imaging velocimetry (PIV) and planar laser-induced fluorescence technique (PLIF). Velocity and CO_2 concentration fields in the liquid after the impingement were visualized. The results show that the impingement of a single liquid drop on the water surface generates several vortex rings near the water surface. The vortex rings renew the water surface and also convect the CO_2 gas dissolved near the water surface downward. The vortical motion clearly shows that the vortex rings work as surface-renewal eddies. The radius, center velocity and presence time of surface-renewal eddies increase with increasing momentum of the impinging drop. This suggests that surface-renewal eddies with larger radius and faster center velocity are induced by the impingement of a single drop with larger vertical momentum, and air–water mass transfer is promoted by such eddies. Based on the surface-renewal concept including the area and time fractions, a model for the air–water mass transfer due to multiple impingements of drops is also proposed.

1. Introduction

Turbulent mass transfer across the air–water interface due to impingement of liquid drops is of great interest in both geophysics and engineering. Particularly, from the geophysical point of view, the estimate of the air–sea mass (CO_2) transfer rate due to impingements of raindrops on the sea surface is expected to improve predictions of future climate change, since the air–sea interface may be broken and mixed by impingements of raindrops. Therefore, it is of great importance to investigate the air–water mass transfer mechanism due to the impingement of a single liquid drop and to investigate the relationship between the mass transfer coefficient due to multiple impingements of drops and the drop concentration.

Mixing of a single drop into quiescent receiving water is not simple because of the water-surface deformation. The splashing, coalescence, bouncing, bubble entrainment, underwater ambient noise and vortex rings generated by single-drop impingement have been the objects of many experimental and numerical investigations (e.g. Worthington, 1908; Harlow and Shannon, 1967; Nystuen, 1986; Rodriguez and Mesler, 1988; Hsiao et al., 1988; Peck and Sigurdson, 1994; Cresswell and Morton, 1995; Buchholz and Sigurdson, 2000; Thoroddsen et al., 2003). The details of the fluid mechanics induced by the impingement of a single liquid drop are well reviewed by Prosperetti and Oguz (1993) and Rein (1993). However, there has been no investigation of the mechanism of air–water mass transfer promoted by the impingement of a single liquid drop.

In addition, for multiple impingements of drops, the mechanism of air–water mass (or heat) transfer due to the drops' impingement has been modeled in some studies (Green and Houk, 1979; Schlüssel et al., 1996; Zappa et al., 2009). Green and Houk (1979) measured the vertical temperature distributions in the thermal mixed-layer generated by the impingement of uniform liquid drops using a multiple-drops generator placed 14 m above a receiving tank attached with 22 thermopiles. They experimentally investigated the relationship between the thickness of the thermal mixed-layer and the drop concentration or mean vertical kinetic energy flux of drops *KEF*. Schlüssel et al. (1996) theoretically proposed a surface-renewal model for evaluating the impact of precipitation on the thermal and diffusive molecular boundary layers of the ocean. But the authors have not clarified the relationship between the mass transfer coefficient and drop concentration in their models. Zappa et al. (2009) measured the mass transfer coefficient k_L enhanced by multiple impingements of drops and turbulent dissipation rate ε through gas exchange experiment at the Biosphere 2 Center. They concluded that k_L can be scaled with $\varepsilon^{1/4}$, even though they measured k_L under only five conditions of the drop concentration. The air–water mass transfer mechanisms due to the multiple impingements of drops has been modeled based on the development of the mixed layer due to multiple impingements of drops

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http://dx.doi.org/10.1016/j.ijmultiphaseflow.2013.11.006

without observations of the vortical motions near the water surface generated by the impingement of a single drop.

The purpose of this study is to clarify the air–water mass-transfer mechanism due to the impingement of a single liquid drop, and to propose a new model for air–water mass transfer due to multiple impingements of drops under the condition of thin drop concentration without interaction between multiple drops.

2. Experiments

2.1. Experimental apparatus and methods

Fig. 1 shows the experimental apparatus. The test section in a receiving tank, made of acrylic plate, was 0.1 m long, 0.1 m wide, and 0.1 m deep. Distilled water was supplied into the receiving tank, and the water depth was 0.06 m. Furthermore distilled water was supplied into a head tank (0.1 m long, 0.1 m wide, and 0.1 m deep) located at 0.2–1.9 m over the receiving tank. A single hypodermic needle was installed at the bottom of the head tank. Two needle diameters, d_n = 0.4 and 0.8 mm, were used here; the drop diameters d_p were 2.2 and 2.8 mm, respectively. In order to generate larger drops of d_p = 4.0 and 5.6 mm, plastic tubes with two diameters, d_t = 2.0 and 8.0 mm, were attached on the needle of d_n = 0.65 mm (see Fig. 2). The vertical distance between the tip of the needle and the free surface of the receiving water (needle elevation, h_t) was changed from 0.167 to 1.867 m.

The falling motion of a single drop was visualized just on the free surface in the receiving tank at several heights of h_t = 0.167–1.867 m by means of a high-speed video system suitable for long-time recording (NAC HSV-500, see Fig. 1). The high-speed video was operated at a speed of 500 frames per second to capture the falling motion of the single drop. The impinging velocity of a drop on the free surface v_p was measured by analyzing the video frames of a falling drop, and the diameter of a drop d_p was estimated from the measured weight of 100 falling drops by assuming a spherical shape. The values of v_p ranged from 1.79 to 5.38 m/s. The relationship between needle elevation h_t and impinging velocity v_p for four liquid drop diameters d_p of 2.2, 2.8, 4.0 and 5.6 mm is shown in Fig. 3. The solid curves indicate the impinging velocity calculated by using the drag coefficient for a sphere C_D proposed by Schiller and Nauman (1933):

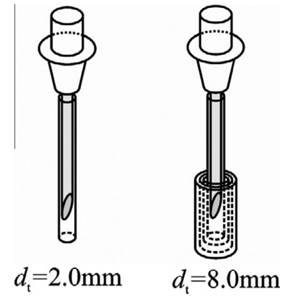

Fig. 2. Sketch of attachment of a tube with outer diameter d_t ranging from 2.0 mm to 8.0 mm.

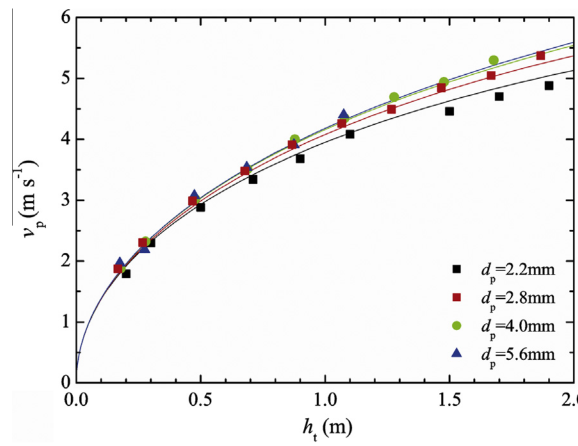

Fig. 3. Relationship between needle elevation h_t and impinging velocity of a drop v_p with the diameter of 2.2 mm, 2.8 mm, 4.0 mm or 5.6 mm. Solid lines denote the numerical predictions by Eq. (4).

$$C_D = \frac{24}{Re_p}(1 + 0.15 Re_p^{0.687}) \quad \text{for} \quad Re_p \leqq 800. \tag{1}$$

In order to consider the effect of the drop's deformation at $Re_p > 800$, C_D were calculated by the following best-fit curve for the data of Gunn and Kinzer (1949):

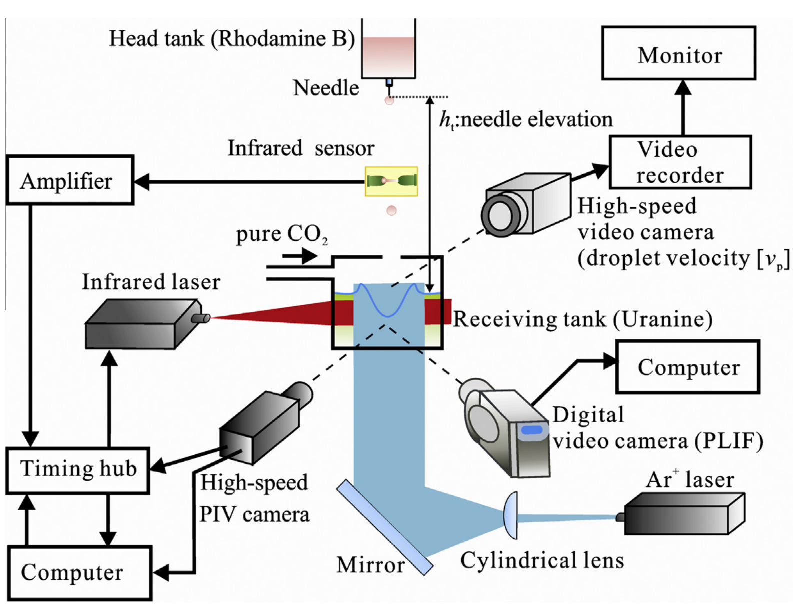

Fig. 1. Experimental apparatus and PIV/PLIF system for measuring velocity and concentration after the impingement of a single liquid drop.